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GaN Epitaxial Growth on Sapphire Substrates by NH_3 Source Molecular Beam Epitaxy

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We have investigated in detail dependence of processes of annealing GaN buffer and GaN growth layers on a substrate temperature using NH_3 gas source molecular beam epitaxy. These processes have been observed by ***in-situ*** reflection high-energy electron diffraction and atomic force microscopy. It is found that there is difference in the surface morphology of epitaxial GaN layer between at growth temperatures below 950 ℃ and 1000 ℃. It has been considered that the growth kinetics of GaN epitaxial layer is extremely changed at 1000 ℃.

Key words: GaN, NH_3, Epitaxy, MBE, RHEED

1. Introduction

The GaN and related compounds are promising materials for blue and short wavelength optoelectronic devices [1-4]. Many GaN-based applications of optoelectronic device are demonstrated on sapphire substrate because sapphire substrate has advantages such as the thermal stability and inactivity for nitridation. However, it is hard to obtain the high-quality GaN on a sapphire substrate with low density of threading dislocation because of the large misfit and the thermal coefficient between GaN and sapphire [5-14]. It is essential for realization of the ideal GaN epitaxial layer to study the initial anneal and growth processes of GaN epitaxial layer at a high growth temperature of 1000 ℃. There has been little work on the ***in-situ*** investigation of annealing and growth processes of GaN at high temperatures using NH_3 gas source molecular beam epitaxy (GS-MBE) [15, 16].

In the present work, we have investigated the thermal annealing process of GaN buffer layer deposited at a temperature of 600 ℃ and initial growth stage of GaN epitaxial layer on sapphire substrate using NH_3 GS-MBE with ***in-situ*** reflection high-energy electron diffraction (RHEED) and ex-situ atomic force microscopy (AFM) observations.

2. Experiment

GaN have been grown by using of a GS-MBE apparatus with an ***in-situ*** RHEED system. Since details of apparatus equipment have been reported previously [9], only a brief outline is described here. A growth chamber was evacuated by a molecular vacuum pump, and the base pressure was 1×10^{-10} Torr. $Al_2O_3(1000)$ wafer was used as substrates and thermally annealed at a substrate temperature of 950 ℃ following chemically etching by dipping in a solution of HF : H_2O = 1 : 50. The sapphire substrate was set on sample holder with Si tips which is rolled as a thermal heater by directly passing a dc electric current. Substrate temperatures were kept at from 600 to 1000 ℃ measured by an optical pyrometer. The emissivity of the pyrometer is adjusted for the temperature of Si substrate using as a heater. NH_3 gas was introduced into the growth chamber through a delivery stainless tube. The pressure of NH_3 gas during GaN growth was controlled by a mass-flow controller in the range of 2×10^{-6} to 2×10^{-4} Torr. The surface structure and film growth behavior of sapphire substrate and GaN epitaxial layer were observed by ***in-situ*** RHEED system. The electron beam was operated at an acceleration voltage of 10 kV. The surface morphology of samples is observed by ex-situ AFM. The diffraction images for sequences were detected with a digital still camera.

3. Results and Discussions

3.1 Thermal cleaning of sapphire

Figure 1 shows a RHEED pattern taken from a sapphire substrate after thermal annealing at 950 ℃. In this figure, sharp and strong diffraction streaks on the zero-order Laue zone are observed. Furthermore, super diffraction streaks are clearly observed, which indicated that the surface reconstructed structure is formed on the substrate. Therefore, it is considered that the surface of sapphire is extremely clean and flat.

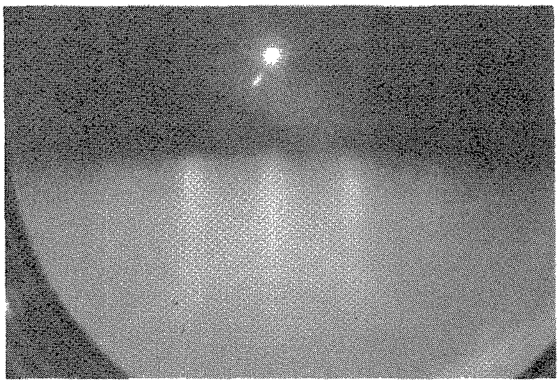

Fig. 1 RHEED pattern from sapphire substrate after thermal annealing at 950 ℃.

3.2 Anneal processes of GaN buffer layer

In previous studies[6], we have reported that it is essential for high quality GaN layer to proceed the annealing of GaN buffer layer at a temperature of above 950 ℃.

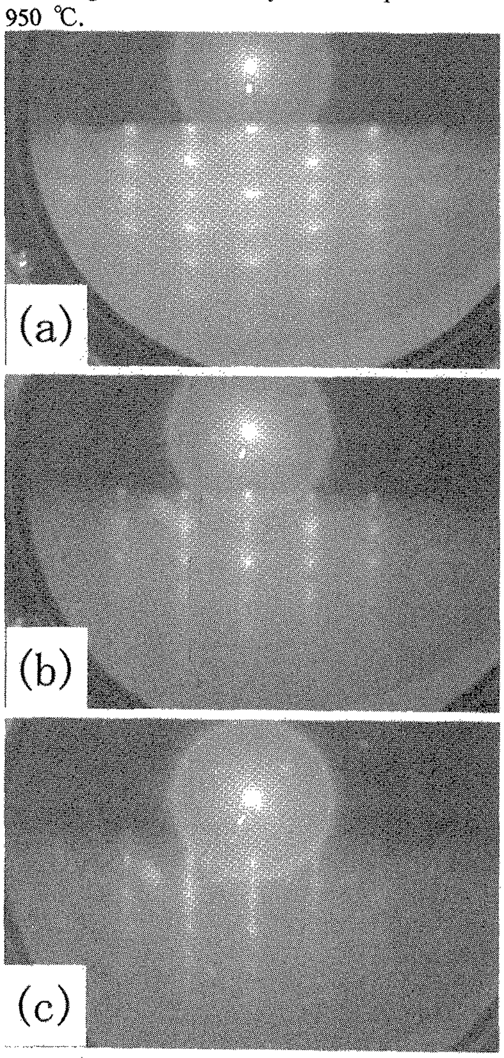

Fig. 2 ***in-situ*** RHEED patterns taken from GaN buffer (a) as-deposited at 600 ℃, (b) thermal annealing at 1000 ℃ for 30 s and (c) after thermal annealing at 1000 ℃ for 60 s.

Figure 2 shows RHEED patterns taken from various steps of GaN buffer layer, (a) as-deposited on Al_2O_3(1000) surface at a temperature of 600 ℃ for 60 min, (b) during thermal annealing at 1000 ℃ for 30 s and (c) after thermal annealing at 1000 ℃ for 60 s. It is observed that RHEED pattern changes from broad spots to sharp streaks as proceeding thermal annealing for only 60 s. In the case of thermal annealing at 950 ℃, it is need for 10 min to obtain the distinctly change of RHEED pattern. This result is consistent with AFM analysis shown in Fig. 3.

Figure 3(a) is in-plane AFM image of GaN buffer layer as-deposited at 600 ℃ for 60 min. It is found that the deposited surface is covered with much number of small GaN islands. Fig. 3 (b) is AFM image for the GaN buffer layer after thermal annealing at 950 ℃ for 10 min. It is found that the shape of GaN islands is changed from small grain to scaled-shaped structure with relatively large size of around 1μm- φ.

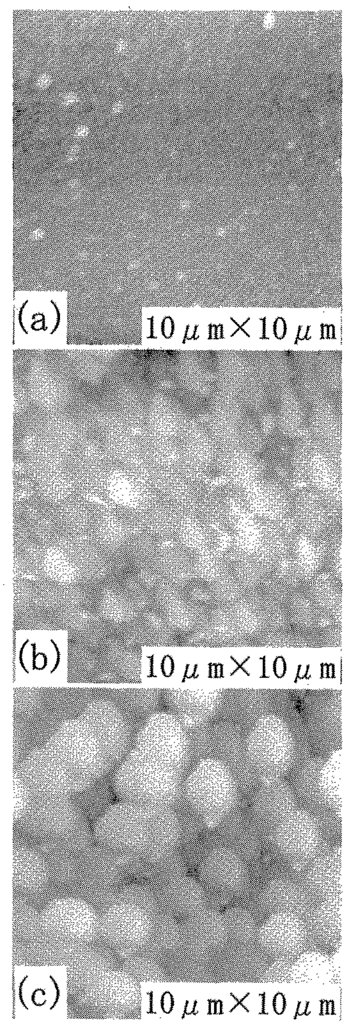

Fig. 3 In-plane AFM images for GaN buffer layers of (a) as-deposited at 600℃, (b) thermal annealing at 950 ℃ for 10 min and (c) thermal annealing at 1000 ℃ for 60 s.

Figure 3(c) shows in-plane AFM image of GaN buffer layer after thermal annealing at 1000℃ for 1 min. It is found that the columnar-shaped structure of GaN islands is appeared in spite of scale-shaped one. Furthermore, the size of these columnar-shaped island is uniform and large of about 2 μm- ϕ .

These results indicate that the crystallization of GaN buffer layer is occurred in the solid phase epitaxy mode by thermal annealing and the structure of annealed GaN layer extremely depend on the annealing temperature.

3.3 Growth processes of GaN at 1000℃

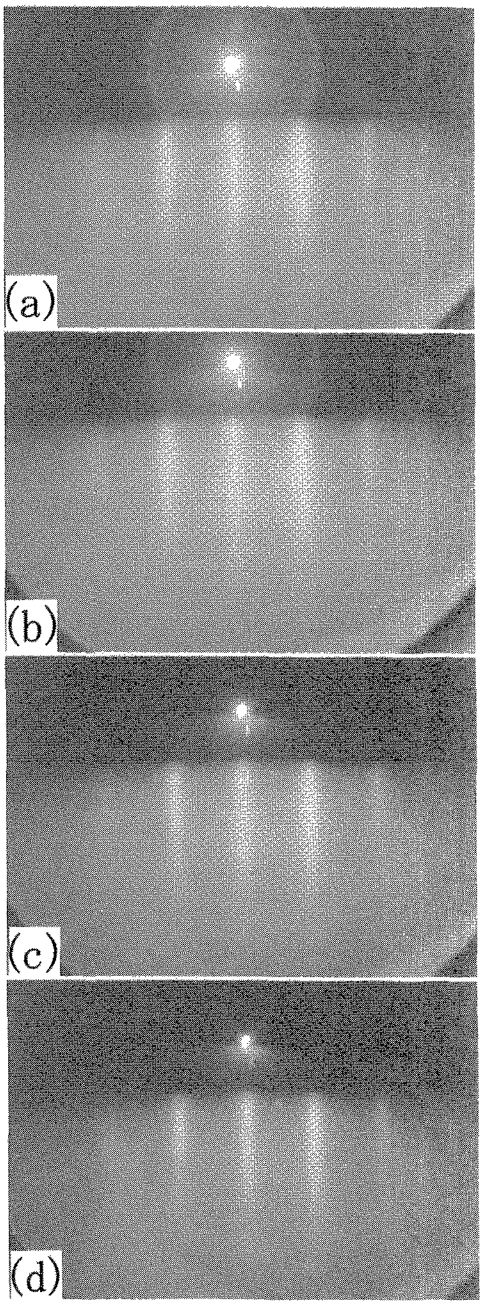

Fig. 4 RHEED patterns GaN epitaxial layer at 1000 ℃ for the growth time of (a) 5, (b) 10, (c) 60 and (d) 120 min, respectively.

GaN epitaxial layer have been grown at 1000℃ on annealed GaN buffer layer. RHEED patterns of fig. 4 are taken from GaN epitaxial layer for the growth time of (a) 5, (b) 10, (c) 60 and (d) 120 min, respectively. In this figure, it is found no change of RHEED pattern through the GaN growth. This result suggests that GaN epitaxial layer is grown in stable mode.

Figure 5 shows in-plane AFM images for GaN epitaxial layer after growth at (a) 950 ℃ and (b) 1000 ℃. It is found that there is larger difference in surface morphology between fig. 5(a) and 5(b). In the case of growth at 950 ℃, the columnar structure with hexagonal shape is observed. On the other hand, continuous film structure is appeared after growth at 1000 ℃.

These results suggest that there are two kinds of growth mechanisms corresponding to two kinds of GaN structure depending on the growth temperature. The kinetic process of GaN epitaxial layer growth can be considered to consist of the various processes of NH_3 adsorption, decomposition of adsorbed species and surface migration of Ga atoms. It is reported that the surface structure is changed between islanding and flat one depending on the polarity factor of the growing surface [10, 11]. In the case of N-polarity layer, the columnar structure is formed on the surface. On the contrary, a flat surface is formed on the Ga-polarity film[11]. Therefore, it is considered that the polarity of GaN epitaxial layer is changed by growth temperature. From these results, it is suggested that high growth temperature over 1000 ℃ is a suitable condition to obtain a high quality GaN epitaxial film. Further analysis of the kinetics of the growth process is in progress to make clear the mechanism of the present GaN epitaxial growth.

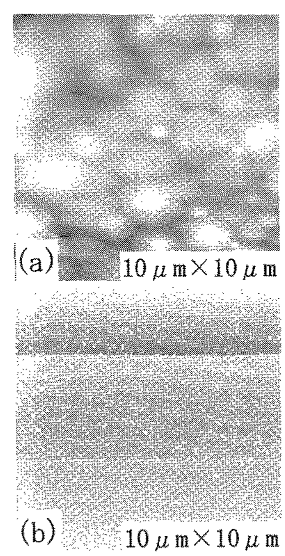

Fig. 5 AFM images for GaN epitaxial layer at a growth temperature of (a) 950 ℃ and (b) 1000 ℃ for the growth time of 120 min.

4. Conclusions

We have investigated in detail dependence of processes of annealing GaN buffer layer and GaN growth on a substrate temperature around 1000 ℃ using NH_3 gas source molecular beam epitaxy. The behavior of these has been observed by ***in-situ*** reflection high energy electron diffraction and atomic force microscopy. It is found that there is difference in the surface morphology of epitaxial GaN layer between below growth temperatures under 950 ℃ and that of 1000℃. It has been considered that the growth kinetics of GaN epitaxial layer is extremely changed at 1000 ℃.

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